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Theory of $He_2^+ + O_2$ Charge Exchange Laser

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July 1977

NAVAL RESEARCH LABORATORY
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The kinetics of O_2^+ first-negative band laser due to near resonance charge exchange between He_2^+ and O_2^- are presented. It can be excited in the afterglow of a discharge in a high pressure He and O_2^- mixture ($He \gg O_2^-$). Gain calculation, however, is presented for discharges due to high energy proton beams advocating their use for high pressure gas lasers. In addition, a comment is made on the disappearance of 3914 laser lines with increasing N_2^- density in the $He_2^+ + N_2^-$ charge exchange laser.

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THEORY OF $He_2^+ + O_2$ CHARGE EXCHANGE LASER

Resonance charge exchange between positive ions and neutrals have been proposed[1] as a mechanism for inversion. A large number of lasing atomic lines have been observed[2-4] to arise as a result of this mechanism, where the charge exchange occurs between an atomic ion and an atom. Furthermore, these lasers have been realized experimentally in low pressure gas discharge systems. More recently, however, lasers have been observed in the afterglow of a high pressure gas discharge[5] in He and N_2, due to a near resonance charge exchange between a molecular ion and a neutral molecule (e.g. $He_2^+ + N_2$). This same laser has also been observed[6,7] by the application of relativistic electron beams incident on a high pressure gaseous mixture.

In this letter we give the kinetics of a new laser in the visible similar to $He_2^+ + N_2$ laser, calculate its gain coefficient and show that it is highly possible. It can be excited by electron or proton beams. We do the calculations using current proton beams (0.3 - 1 MeV) by pointing out[8] that these beams produce more ion pairs compared with electron beams of the same energy. Consequently higher laser power densities are derivable[8] using proton beams compared with electron beams.

The new laser under consideration arises from the near resonance charge transfer between He_2^+ and O_2 according to

$$He_2^+ + O_2 \rightarrow O_2^+(b^4\Sigma) + 2\ He\ . \qquad (1)$$

Emissions due to this process have been observed[9] long time ago in the afterglow of a discharge.

Figure 1 shows the relevant energy diagram[10] of O_2, O_2^+ and the energy positions[11] of He_n^+ ions. This figure shows the near resonance nature of

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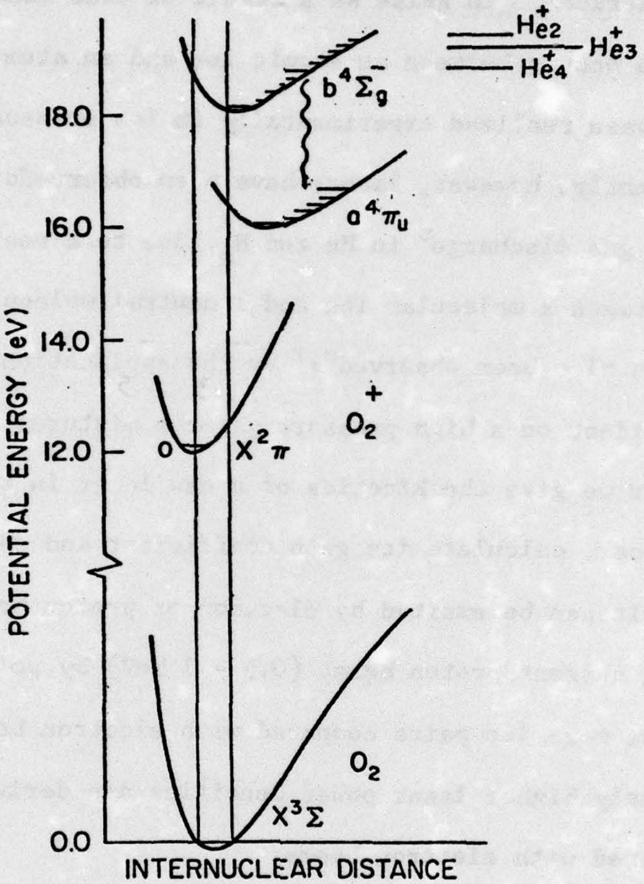

Fig. 1 — The potential energy diagrams of relevant O_2, O_2^+ and energy positions of He_n^+ are shown indicating the near resonance nature of $He_n^+ + O_2$ charge exchange

reaction (1) where higher vibrational levels of $b^4\Sigma$ are favored according to the Franck-Condon principle (vertical transitions, classically speaking). The $b^4\Sigma$ electronic state is the upper level of the first negative bands system which corresponds to $b^4\Sigma \rightarrow a^4\Pi$ transitions in the visible whose strong[12] vibrational bands emission are given in Table 1.

Table 1

λ	I	v′, v″	λ	I	v′, v″	λ	I	v′, v″
7891		0, 4	5833.4	8	3, 3	5295.7	9	2, 0
7348		0, 3	5847.3	2	4, 4	5274.7	10	3, 1
6856.3	9	0, 2	5814	1	5, 5	5259	6	4, 2
6418.7	10	0, 1	5631.9	10	1, 0	5251	10	5, 3
6351.0	10	1, 2	5597.5	10	2, 1	5241	8	6, 4
6026.4	10	0, 0	5566.6	6	3, 2	5234	9	7, 5
5973.4	10	1, 1	5540.7	2	4, 3	5005.6	2	3, 0
5925.6	9	2, 2	5521	2	5, 4	4998	2	4, 1
						4992	2	5, 2

This laser can be developed by the application of an electron or a proton beam incident on a high pressure mixture of He and O_2. These energetic beams create the atomic ion, He^+, which in turn is transformed into He_2^+ according to

$$He^+ + 2\,He \rightarrow He_2^+ + He\,. \tag{2}$$

The He_2^+ ions then charge transfer with O_2 where the total rate coefficient[13] is 10^{-9} cm^3/sec. This includes[13] the dissociative charge exchange with O_2 and therefore one can assume that at least half of the charge exchange is in the form of reaction (1) leading to the upper laser levels. Reaction (2) has a rate coefficient[14] of 1.0×10^{-31} cm^6/sec at 300 °K, however, if the gas temperature is cooled below 300 °K, heavier helium molecular ions are formed and at a faster rate.[15] It is of interest to note, that He_4^+ ions (see Fig. 1) will preferentially excite v = 5, 6 vibrational levels of O_2^+, which leads to green lasers around (5234 - 5259 Å).

To calculate the gain coefficient we consider 0.3 MeV proton beam,[16] current density of 0.1 kA/cm^2 and a pulse duration of 5 nsec incident on 2 atmospheres of He and 2 torr of O_2. A simple analysis can be made including a discussion of the kinetics involved. The number of $\dot{He}^+$ ions, $N(\dot{He}^+)$, formed can be obtained from

$$N(\dot{He}^+) \simeq N(He) \frac{L(E)}{W} N_p V_p \Delta t \; , \tag{3}$$

where N(He) is the density of helium atom, N_p and V_p are the density and the velocity of the incident protons, respectively. L(E) is the stopping power of He for protons, shown in Fig. 2, where calculated and experimental data[17] are indicated. W is the energy expended per ion pair and is[17] ~ 46 eV for He. With the given parameters one obtains $N(\dot{He}^+) = 4 \times 10^{15}$ cm^{-3} in 5 nsec. These ions are transformed into He_2^+ and are neutralized via the collisional radiative recombination. In the next 5 nsec 1.5×10^{15} cm^{-3} He_2^+ ions are formed while only $\simeq 10^{12}$ cm^{-3} $\dot{He}^+$ ions have recombined[18] with electrons, assuming[19] an electron temperature of 0.5 eV. The molecular ions

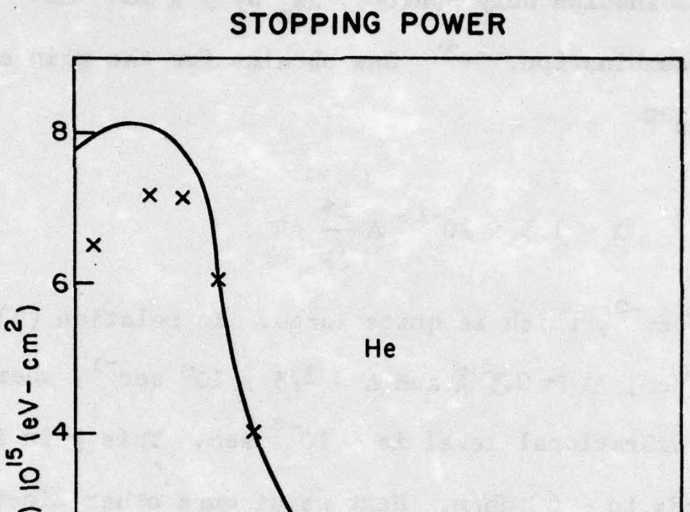

Fig. 2 — The stopping power of He for protons as a function of proton energy. Calculated and experimental (indicated by xxx) values are shown.

charge transfer with O_2 producing 2.6×10^{14} cm^{-3} upper laser level, in 5 nsec. This is considered as the inversion density. During this period 2.6×10^{14} cm^{-3} He_2^+ ions have also dissociatively charge exchanged with O_2, while electron recombination only depletes He_2^+ by 3×10^{11} cm^{-3} via collisional radiative recombination.[20,21] One obtains for the gain coefficient using the expression[22]

$$\alpha = 1.3 \times 10^{-12} A \frac{\lambda^4}{\Delta\lambda} \Delta N , \qquad (4)$$

a value of $\alpha \simeq 0.27$ cm^{-3}, which is quite large. In relation (4) we have used $\lambda = 5.2 \times 10^{-5}$ cm, $\Delta\lambda \simeq 0.3$ Å and $A = 1/3 \times 10^6$ sec^{-1}, where generally the life-time[23] of the vibrational level is $\sim 10^{-6}$ sec. This gain is quite large and corresponds to ~ 60 db/m. Next we discuss other kinetics which occur in the system and evaluate their influence on the laser output. There are three processes which affect the upper and the lower laser levels. These are the dissociative recombination, the quenching by O_2 and the de-excitation by free electrons. If one assumes that the excited states of O_2^+ dissociatively recombine with electrons at the same rate[24] its ground state does, then the upper laser level is reduced by 1.5×10^{14} cm^{-3}. The quenching by O_2 is not known, however, the lower laser level, $a^4\Pi$, is quenched by O_2 with a rate coefficient[25] of 3×10^{-10} cm^3/sec. Assuming a similar rate for the upper laser level, the quenching will reduce the inversion density by $\sim 3 \times 10^{13}$ cm^{-3}. The electron de-excitation rate of $b^4\Sigma(v)$ states can be obtained by analogy with excitations and de-excitation of atoms and ions. Using rates given by Von-Regemorter[26] with a Gaunt factor of 0.2 and an oscillator strength of 0.004, one needs an electron density $\sim 10^{16}$ cm^{-3} to

de-excite the upper laser levels at a rate equal to their radiative decay rates. At an electron density of 4×10^{15}, the de-excitation rate is still below the neutral quenching rate. Thus, the total inversion density is reduced to $\sim 8 \times 10^{13}$ cm^3/sec giving a gain coefficient of $0.08\ cm^{-1}$. However, if the electron temperature is close to 1 eV then the inversion density will be $\sim 1.3 \times 10^{14}\ cm^{-3}$ giving a gain coefficient of $0.13\ cm^{-1}$. These calculations show the strong possibility of a successful development of such a laser.

In addition to these above processes one may consider other reactions which also occur and try to estimate their effects on the inversion density. One such process is the formation of the negative ion, O_2^-. The formation rate of O_2^- when the third-body is N_2 has a rate coefficient of $\sim 10^{-31}$ cm^6/sec. Using this rate for our mixture one obtain $O_2^- \simeq 10^{12}\ cm^{-3}$ whose influence on the upper laser level and He_2^+ in terms of their mutual neutralization is negligible. Finally the formation of O_4^+ via the depletion of the $(O_2^+)^*$ is also negligible, since the three-body rate coefficient for such a process is[27] $\sim 2.8 \times 10^{-30}$ cm^6/sec.

Finally, increasing partial pressure of O_2 should terminate the laser power output due to quenching. Speaking of quenching we would like, at this juncture to make the following comment relevant to another laser. In the $He_2^+ + N_2$ charge exchange laser, obtained by electron beam pumping[6,7,28] and recently in a regular electric discharge,[5] one observes the disappearance of the (0, 0) band at 3914 Å, with increasing partial pressure of N_2. No physical explanation has yet been offered for these observations. We would like to suggest that the disappearance of the 3914 Å with increasing partial pressure of N_2 is due to the quenching of $B^2\Sigma(v = 0)$ state by N_2. The

quenching rate coefficient[29] is 4×10^{-10} cm^3/sec. Thus at N_2 = 10 torr, e.g., the quenching rate is 1.4×10^8 sec^{-1} compared with the total radiative decay rate[30] of 1.58×10^7 sec^{-1}. The total radiative decay rate of $B^2\Sigma(v = 0)$ consists of decays of (0, 0), (0, 1) and (0, 2) bands which are[30] 1.24×10^7 sec^{-1}, 2.2×10^6 sec^{-1} and 5×10^5 sec^{-1}, respectively. The relative rates of these bands are 0.82: 0.14: 0.03, and in principle the quenching of $B^2\Sigma(v = 0)$ state by N_2 should follow these ratios as well. Obviously the 3914 disappears with increasing N_2 partial pressure, because it is quenched much faster then the other transitions which require much more higher N_2 pressures to be quenched. The power output at 3914 Å is still very high,[31] however, the duration, which is N_2 dependent, is very short requiring special means for detection (especially for $\Delta t \leqslant 1$ nsec).

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